Greening Coal: Breakthroughs and Challenges in Carbon Capture and Storage
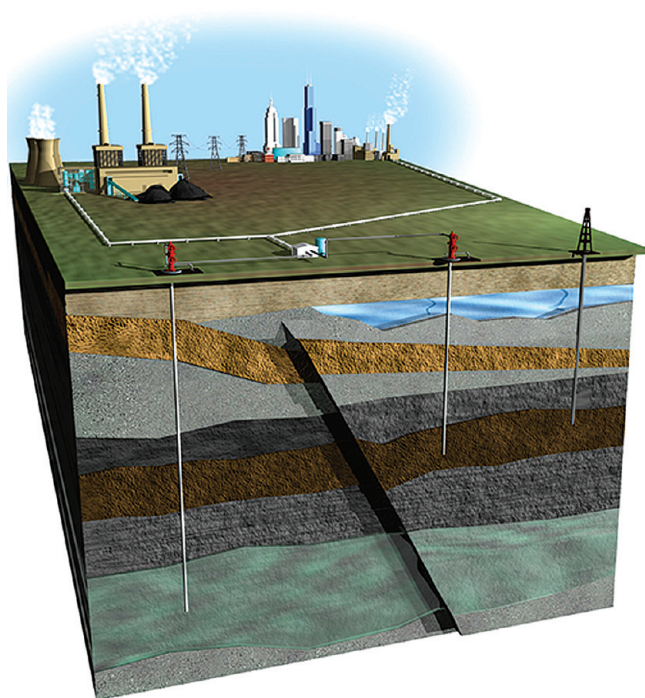
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INTRODUCTION

Carbon capture and storage (CCS) is a critical technology for reducing emissions of greenhouse gases (GHGs) to the atmosphere. CCS is being considered as one piece of a strategy for stabilizing atmospheric CO_2 concentrations.[1] This plan requires that globally, billions of tonnes of carbon dioxide ($GtCO_2$) each year must be captured, concentrated, and stored to keep it out of the atmosphere for hundreds to thousands of years. The near-term approach is to capture and compress CO_2 from stationary industrial sources (e.g., coal and natural gas burning power plants) and transport it through pipelines for injection and long-term storage in geologic reservoirs (e.g., depleted oil/gas fields and deep saline aquifers). In 2009, U.S. coal power plants generated 307 gigawatts of electricity (GWe) and produced 2.4 $GtCO_2$ out of total U.S. emissions of 6 $GtCO_2$.[2] The existing fleet of coal-fired power plants will continue to be a major source of electricity for the next 20 years, with estimated production capacity increasing to 400 GWe.[3] In addition, electrical generation in China has expanded rapidly in recent years, nearing the size of the U.S. fleet,[4] and three-quarters of China's power plants burn coal.[5] Given the persistence of this global capacity for coal combustion for the next few decades, CCS represents a bridging technology that will allow us to continue to generate electricity in existing power plants while we transition to a low-carbon energy future.

CCS technology must be deployed at a massive scale to have a meaningful impact on reducing industrial CO_2 emissions to the atmosphere. This could require the U.S. to capture on the order of 1 $GtCO_2$/yr from hundreds of typical coal-burning power plants and to construct dedicated pipelines to handle a CO_2 volume 25% greater than the U.S.' 2009 daily oil consumption.[6] Additionally, this volume of CO_2 will require finding extensive geologic formations in low risk environments to store between 1 - 3 km^3 of supercritical CO_2 each year.

This paper explores the science and technology related to CO_2 capture, geologic storage, and system-wide integration. We emphasize strategies and technologies suitable for making CCS a reality in the near future, with particular focus on retrofitting existing coal-fired power plants to capture and compress CO_2 for geologic storage. Projects involving coal combustion retrofits currently represent an area of particular focus for implementing CCS in the power industry in the next decade. Examples include turbine retrofit and capture of 1 million tonnes per year ($MtCO_2$/yr) at the coal-fired Boundary Dam plant (SaskPower) in Saskatchewan,[7] oxy-fuel combustion retrofit and capture of 1.3 $MtCO_2$/yr in the FutureGen 2.0 plant (Ameren) in Illinois,[8] and postcombustion capture of 3,000 tCO_2/yr at the Gaobeidian power plant in Shanghai.[9]

CO₂ CAPTURE

Power plants are responsible for greater than one-third of the CO_2 emissions worldwide and are a prime focus of global CCS efforts.[10] Capturing CO_2 from the mixed-gas streams produced during power generation is a first and critical step for CCS. Three strategies for incorporating capture into power generation scenarios are of primary focus today: post-, pre- and oxy-combustion capture (Figure 1).

Postcombustion capture systems are designed to separate CO_2 from pulverized coal (PC) derived flue gas. PC flue gas contains 10—13% CO_2 with a balance of N_2, steam, and other impurities (SO_x, NO_x, heavy metals).[11] Oxy-combustion power plants are modified versions of conventional PC plants using
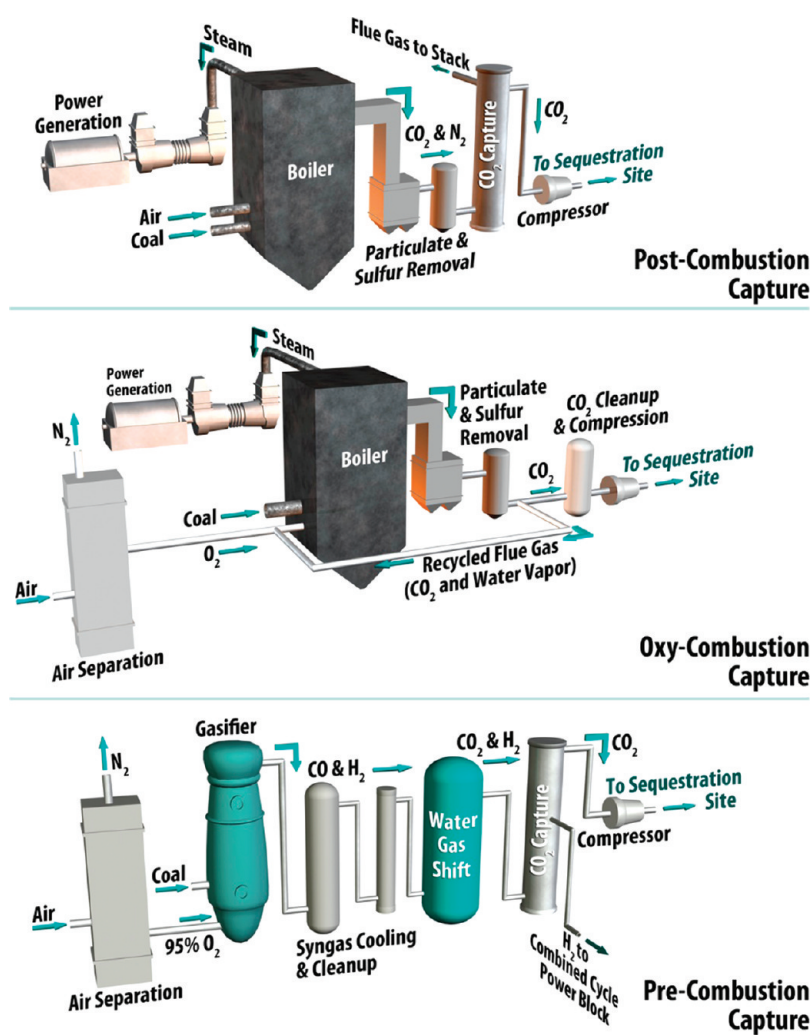
Figure 1. Post-, oxy-, and precombustion concepts and separation system integration into power plants.

oxygen (O_2) diluted with recycled flue gas instead of air to combust coal into steam and high purity CO_2. Precombustion systems are designed to separate CO_2 from synthesis gas ("syngas") prior to electricity/hydrogen (H_2) production and are applicable to new, more efficient, integrated gasification combined cycle (IGCC) plants. Syngas from the coal gasifier is primarily comprised of H_2 and carbon monoxide (CO). A water-gas-shift reactor is added in the capture process to convert CO to CO_2, thus facilitating capture while producing additional hydrogen. Power generation with capture using oxy- and pre-combustion processes has 10−37% higher net efficiency than that of a new air-fired PC plant without CO_2 capture and allows more flexibility for future improvements in design.[12]

The U.S. Department of Energy (DOE) CCS target is to achieve 90% CO_2 capture while limiting the increase in cost of electricity (COE) to 35 and 10%, respectively, for plants implementing postcombustion and pre/oxy-combustion capture.[13] The energy consumption and losses associated with CO_2 capture using today's technologies represent an unacceptably high proportion (>75%) of the total cost of CCS (capture/compression, transport, storage). The DOE cost targets require significant improvements in large scale deployable capture technologies. Although all of these technologies will

be vital in the long term, current U.S. and global power production industries are dominated by PC-based plants. Therefore, postcombustion capture is the most likely to have the largest impact on total CO_2 emissions reductions over the next few decades.

The U.S. Energy Information Administration estimates that, in 2030, 78% of the CO_2 emissions resulting from U.S. electricity generation will still be derived from the current fleet of PC plants.[14] Thus, to effect change in the near term, we believe that these PC emissions must be addressed to a large extent through postcombustion separation and capture retrofits to those plants. CO_2 separation technologies are readily available and have been used industrially for nearly 60 years. These technologies are based on chemical solvents (e.g., monoethanolamine, MEA) and physical solvents (e.g., glycol or methanol). A regeneration step is used to reclaim the solvent for reuse. Although technically suited for CO_2 capture applications, implementation is hindered by exorbitant operating costs due to high energy penalties for solvent regeneration and material and environmental costs due to solvent attrition. In one study, the estimated cost of implementing these existing capture technologies in the operational U.S. power plants will increase the COE by 330%.[13]

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The development of new and innovative capture systems for PC application is imperative. Fortunately, increasing research budgets have enabled scientists and engineers worldwide to make progress in this burgeoning field. Emerging CO_2 separation technologies under various stages of development include solvents, sorbents, membranes, and biologically mediated separation systems. A brief summary describing the important aspects of these emerging technologies related to CO_2 capture from coal-derived power production is provided below.

Technology development goals for improved solvents include the realization of low-cost, noncorrosive, stable, low-toxicity materials with high CO_2 capacity, rapid mass transfer kinetics, low regeneration energy, and high impurity tolerance.[12,13,15−17] Aqueous ammonia (AA) and ionic liquid (IL) based materials are forerunners in current solvent research, development, and demonstration (RD&D) efforts. Systems such as these provide an opportunity for use of less corrosive, more stable solvents with chemically tunable mass transfer rates and capacities, thereby addressing some of the limitations of the more conventional amine-based materials. AA technologies are under pilot-plant and midscale demonstration while IL based solvents are currently at the laboratory development stage.

Solid sorbents work by adsorbing gaseous CO_2 onto a surface, followed by temperature or pressure driven desorption. CO_2 interacts with the sorbent chemically (e.g., immobilized amines and carbonates) or physically (e.g., high surface area metal organic frameworks and zeolites).[12,13,15−17] Since these CO_2-sorbent interactions are weaker than those between CO_2 and chemical solvents, less heat is typically required for regeneration and CO_2 release.[15] Preliminary cost analyses indicate 15% improvement in COE using sorbents as compared to MEA capture.[18] Process and materials optimization remain challenges for this technology. As with sorbents, materials cost, stability, CO_2 capacity and mass transfer optimization are critical to commercial viability. Additionally, movement of large volumes of solids (e.g., fluidized beds), and the mechanical and thermal robustness required for such process schemes, provide additional process, and materials challenges.

Membranes are currently top contenders among new post-combustion CO_2 separation technologies due to their low energy consumption, lack of moving parts, and modular design opportunities. However, they incur extra cost due to flue gas compression required to create the driving force for transport, post cleaning, and/or multistage operation; membranes must also be stable in the presence of flue gas contaminants and high temperatures. Both organic and inorganic membrane approaches are being pursued. Development is currently focused on achieving high CO_2 throughput with adequate selectivity using low-cost materials. The RD&D performance of organic membranes has the potential to reduce capture costs to as low as $23/tCO_2, significantly lower than the $54/tCO_2 cost using existing industrial amine based separation technologies.[19,20] Further development and pilot-scale efforts are ongoing to fully quantify and increase the estimated cost saving of using membranes for CO_2 capture. New efforts employing mechanically robust room-temperature IL based membranes hold promise for realizing unprecedented CO_2 throughputs and capture costs of <$20/tCO_2.[21,22]

As we transition to a younger and more efficient power production fleet, oxy- and precombustion technologies will play a greater role. To be prepared for this transition, we must continue to work now to develop technologies that will support the implementation of these next-generation fossil fuel-driven plants. Oxy-combustion provides an opportunity for near-complete capture; however, this process requires a large-scale air separation unit to produce high-purity O_2. Although cryogenic air separation is a well-established method for O_2 production, it is both capital and energy intensive.[23] Several novel air separation technologies currently under development have the potential to reduce this cost. Engineering studies indicate that these new technologies can reduce the power associated with O_2 production by 70−80%.[12] By example, incorporation of an ion transport membrane for O_2 production is estimated to reduce total plant costs by more than $130/kW over the same plant with cryogenic separation technologies.[24]

IGCC with precombustion capture has the potential to lower power production costs over conventional PC-based power production with capture. Plant efficiencies for IGCC power plants with capture are estimated to be 6% greater than those of a conventional PC power plant with capture.[25] The extent of the predicted efficiency gains and associated cost differentials is dependent on numerous factors including coal type, plant design, and plant location.[25−27] However, neither the technology advantages of IGCC power production nor the DOE CCS targets are fully realized using current, commercially available CO_2 separation technologies. Globally, advanced precombustion capture technologies based on solid sorbents, membranes, and advanced solvents are currently all at demonstration stages from laboratory to full scale. High temperature membranes and sorbents with separation conditions (temperature and pressure) matched to IGCC process conditions are emerging as promising routes to efficient H_2/CO_2 separation.[13,16,17] In addition to gains from capture technologies, advances continue to be made in other process areas such as turbines and O_2 production. The incorporation of these capture and other process enhancements into advanced IGCC plant designs shows promise for additional efficiency gains in excess of 9% over that of the baseline IGCC case referenced above. Realization of these developing technologies at the commercial scale would thus enable realization of the DOE goals of 90% capture with a <10% increase in COE.[24]

■ CO_2 STORAGE

Once captured and compressed, CO_2 must be stored or sequestered for hundreds or thousands of years. Several storage options are available, but storing or sequestering CO_2 deep in the earth is the only technology that has been established on scales large enough to be useful in our quest to divert billions of tonnes of CO_2 from the atmosphere each year.[28] Geologic storage has already been demonstrated, without major incident, by the oil industry for nearly 40 years. The oil industry uses CO_2 to lower the viscosity of subsurface oil and thus enhance oil recovery (EOR) in depleted oil fields. Not all the CO_2 injected is recovered, thus CO_2 EOR projects are de facto CO_2 storage sites. For example, the SACROC facility in West Texas is estimated to have stored approximately 55 $MtCO_2$ since the early 1970s.[29] Additionally, more than 7500 km of CO_2 pipelines currently operating in the U.S. demonstrate that the technology to move large quantities of CO_2 is safe.[30] However, the scale of EOR in the U.S. is 2 orders of magnitude smaller than the total U.S. CO_2 emissions.[31]

Over the past decade, governments around the world have teamed with industrial partners to initiate the first large pilot tests (>1 $MtCO_2$/yr) of geologic sequestration in or near oil/gas

operations (e.g., In Salah and Sleipner). Geologic sequestration of CO_2 has initially targeted oil and gas reservoirs because they have existing infrastructure, operators have intimate knowledge of reservoir performance, and naturally occurring CO_2 being produced as a waste stream near these sites provides a ready source for injection.[32]

However, the total storage in existing oil and gas formations is relatively small, and the proximity of these reservoirs to CO_2 sources is not optimal. For example, CO_2 from sources in the Illinois Basin greatly exceed the capacity of depleted oil and gas reservoirs in this region, leading current research in the direction of using deep saline aquifers to provide the required storage volumes.[33] Deep saline aquifers are nearly ubiquitous worldwide, with an estimated potential storage volume up to 10 000 $GtCO_2$. In addition, deep saline waters typically do not meet drinking-water standards (10 000 ppm TDS) and thus will not be negatively impacted by CO_2 mixing.[34] Deep saline injection is currently planned for the Mt. Simon sandstone in the Illinois Basin in the U.S. as part of the FutureGen 2.0 project. Other proposed geologic storage formations include off-shore sediments, basalt flows, and unmineable coal seams.

To understand how CO_2 is trapped in the subsurface, numerous coupled processes must be considered. Primary trapping mechanisms include *structural trapping* (separate phase, buoyant CO_2 is trapped by a geologic structure), *residual trapping* (discontinuous CO_2 bubbles are trapped after the bulk of the CO_2 flows through a system), *solubility trapping* (CO_2 dissolves into brine or oil), and *mineral trapping* (CO_2 leads to formation of minerals in the pore space of the rocks). Structural and residual trapping are likely to be the dominant processes on a scale of tens to hundreds of years, while mineralization and convective mixing operate over longer time scales.[35,36] The physical processes controlling these mechanisms depend on many parameters, including temperature, pressure, salinity, pore structure, and capillary forces.

Several of these same coupled processes can negatively impact geologic sequestration systems. Geo-mechanical impacts can be caused by changes in pressure and temperature. First, lowered effective confining stresses caused by increased pore pressure can allow existing faults to reactivate and cause CO_2 to migrate upward. Second, thermal contraction caused by changes in temperature as cool CO_2 is injected into hot rock can damage rocks around the injection location and cause wellbores to fail. Injected CO_2 may also affect the chemistry of deep saline reservoirs, resulting in the formation of minerals that can reduce permeability or dissolution of minerals that can increase permeability. Near injection wells, crystalline salt forms in rock pores due to evaporation of water into supercritical CO_2 and can lead to reduced injectivity of CO_2.

To incorporate this level of complexity, state-of-the-art process level simulators now include thermo-hydro-chemical-mechanical (THCM) processes. Although subsets of these coupled processes have been investigated previously for other applications, mechanistic coupling of more than two coupled processes has not been investigated extensively and represents an exciting research frontier.[37] Input to these numerical models comes from experiments, pilot studies, and natural analogue sites where CO_2 has been stored in geologic reservoirs for tens of thousands of years.[38] For example, measurements of element solubility in the presence of CO_2 in the laboratory and in the field provide modelers with targets to ensure that models can reproduce field data.[39,40] Calculations of complex coupled processes provide the basis for reduced-complexity algorithms that are being used in Monte Carlo risk assessment modeling of sequestration systems with potentially dozens of uncertain parameters.[31,41,42]

Teams around the world are working to assess risk for geologic storage sites. Such risk assessments are vital to the success of geologic sequestration and provide regulators and citizens with confidence in site selection and performance goals.[43] Performance metrics for risk include aspects of health and human safety, environmental degradation, economic impacts, and project planning. In addition to risk assessment undertaken by industry (e.g., CO_2 Capture Project[44]), three major efforts are being coordinated by the International Energy Agency, the U.S. National Risk Assessment Program and the Canadian International Performance Assessment Centre for Geologic Storage of CO_2. Primary risks identified by these groups include leakage of brine and CO_2 from the storage reservoir into overlying aquifers or other subsurface resources (e.g., contamination of oil fields), induced seismicity triggering damaging earthquakes (e.g., a recently abandoned geothermal project in Switzerland[45]), lack of injectivity that severely limits the capacity of proposed storage reservoirs, leakage back into the atmosphere on a scale large enough to negate the sequestration effort (greater than 0.01% per year[46]), and point-source leaks to the surface that could impact human health. These risks are characterized by a combination of high consequences with generally low probability. Many of these risks can be reduced through a combination of site selection, reservoir pressure management, site data collection, and long-term monitoring with contingency plans.[47−50]

Recent developments from analysis of the deep, permeable sedimentary basins of western Wyoming highlight the interplay between the complex costs and benefits associated with CCS. For example, the Rock Springs Uplift (RSU) is currently being considered as a target reservoir for large scale CO_2 sequestration (approximately 26 $GtCO_2$ capacity[51]), and modeling results show that reservoir management must be considered as an integral part of a total sequestration project to minimize risk of leakage. During reservoir management, brine can be removed (produced) from the reservoir to reduce pressure. The volume of brine production required to reduce seismic and leakage risks to near zero is approximately equal to the volume of injected CO_2.[47] For the Jim Bridger power plant near the RSU, nearly a cubic kilometer of CO_2 could be injected over 50 years, leading to a cubic kilometer of produced brine.[52] Produced brines at the RSU will arrive at the surface at temperatures in excess of 100 °C and are a potentially valuable thermal energy resource. The brine can also be desalinated; representing a potential source of freshwater in the arid western U.S. Coproduction of brine is likely to be an attractive resource associated with CO_2 injection in many other parts of the world, including the Ordos Basin in China, currently under study by the joint U.S.-China Advanced Coal Technology Consortium.

■ SCALING UP: MAKING CCS A REALITY

National goals for CO_2 management will require CCS technology to be deployed in a very different manner than existing energy infrastructure. Oil, natural gas, and electricity transmission infrastructures expanded in an ad hoc and incremental fashion over many decades, primarily in response to emerging markets, resource discoveries, and population growth. In contrast, CCS will require a coherent development strategy that considers the intersection of policy, science, and industry.
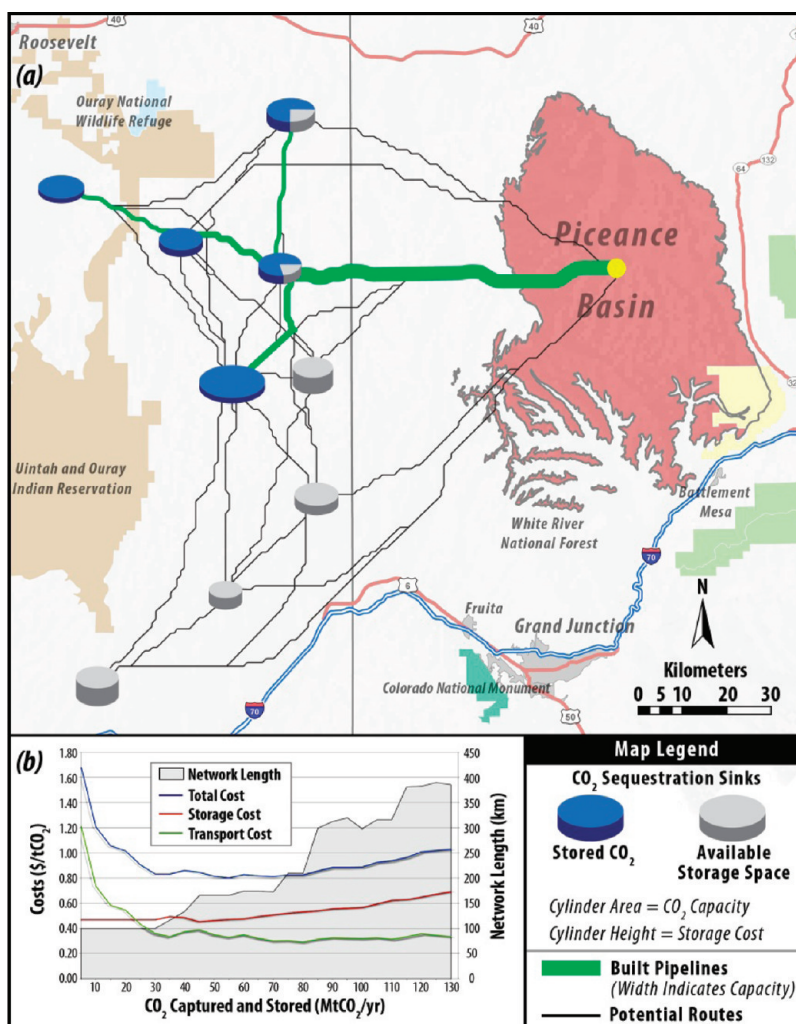
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Figure 2. (a) *SimCCS* infrastructure required to transport/store 80 MtCO$_2$/yr from an oil shale industry. (b) Costs to transport/store CO$_2$ and network length for 26 different CO$_2$ management scenarios (5–130 MtCO$_2$/yr).

Further, CCS technology will have to be applied over the next two or three decades to have a significant impact. Investing in infrastructure at this scale will necessitate careful and comprehensive planning. Specifically, decisions will need to be made regarding where, how much, and with what technology to capture CO$_2$; where and how much CO$_2$ to store in geologic reservoirs; where and what capacity pipelines to construct; and how to allocate CO$_2$ between numerous CO$_2$ sources and sinks. Thus, CCS integration may best develop at the basin scale to make best use of natural resources, clustering of industry and population centers, and topography.[53] These integrated networks will realize economies of scale and proper infrastructure placement that will keep costs down and minimize environmental issues such as residential exposure to CO$_2$ hazards.

Numerous models have been developed to understand how an integrated CCS system—CO$_2$ capture, transport, and storage infrastructure—could and should be deployed. Early CCS integration models focused on straightforward source-sink matching, typically connecting CO$_2$ sources directly to their closest geologic reservoir or sink. These models relied on simplifying assumptions, including geographically impervious straight pipelines,[54] that all CO$_2$ must be captured from a source regardless of system-wide economics,[55] and that pipelines cannot be networked

to aggregate CO$_2$ flows.[56] These early approaches paved the way for more comprehensive CCS models that take into account detailed economics coupled with spatially realistic pipeline networks. For example, a carbon management decision support system, *SimCCS*,[57] simultaneously optimizes the financial investments, operational costs, system capacities, and geospatial construction of CCS infrastructure, as well as routing and networking pipelines across a real-world cost surface. Coupling reservoir performance and risk assessment models (e.g., CO$_2$-PENS,[31,41]) with system optimization models (e.g.,[53,57]) provides a more coherent understanding of the effects of reservoir capacity and costs as they relate to optimal pipeline network design (Figure 2), and the propagation of uncertainty through the CCS system.[6]

Following the more comprehensive approach taken by *SimCCS*, infrastructure models have evolved to address other sophisticated and critical aspects of infrastructure modeling. These state-of-the-art modeling techniques illustrate how CCS infrastructure may evolve in response to a range of policy decisions. For example, Morbee et al.[58] extended the *SimCCS* optimization to allow infrastructure to be constructed gradually through time as the amount of CO$_2$ to manage varies (e.g., cap-and-trade). Kuby et al.[59] contrast optimal CCS systems deployed in response to a CO$_2$ tax versus a cap-and-trade environment. The effect of introducing CO$_2$

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certificates or permits on CCS technology in Europe has been explored by Mendelevitch et al.:[60] higher priced certificates result in greater adoption of CCS technology and reduce CO_2 emissions. CCS deployment in The Netherlands has been mapped out by Van den Broek et al.[61] using a GIS-based optimization energy model; study results could help policy makers reduce CO_2 emissions by as much as 50% of 1990 levels by 2050. These next-generation models will be used by industry to make informed decisions about managing CO_2. For example, the Alberta tar sand oil industry, projected to produce as much as 108 $MtCO_2$/yr by 2020,[62] is planning an integrated network to aggregate CO_2 from multiple locations into one or more CO_2 reservoirs.[63] This network could reduce Alberta's CO_2 emissions by 35 $MtCO_2$/yr by the mid 2020s and perhaps as much as 100 $MtCO_2$/yr in the long term. An analysis of the CO_2 management requirements for a mature U.S. shale oil industry, producing 1.5 million barrels of oil a day, concludes that an integrated pipeline network is the only feasible approach.[53]

Currently, the cost of CO_2 capture ca. \$54/tCO_2,[19,20] amounts to the greatest share of the cost of CCS integration, compared to the cost of transport and geologic storage (<\$10/tCO_2 [53,64]). However, as new capture technologies come online and CO_2 transport and storage networks are developed at the regional scale, these component costs are expected to converge. Although CCS development at the national scale is unlikely to be optimally designed, there are emerging examples of large-scale infrastructure integration. For instance, Southern Company is planning to implement carbon management across its fleet of coal power plants, using existing natural gas pipeline rights-of-way to design CO_2 transport and storage infrastructure in advance of national carbon pricing (or tax) policies. This CCS integration is part of a business model of vertical integration within the utility.[65] In addition, Denbury is converting natural gas lines and building new pipelines for CO_2 to expand connectivity among CO_2 sources (natural and anthropogenic) and EOR projects along the U.S. Gulf coast.[66]

CONCLUDING REMARKS

CCS is a critical technology for reducing near-term CO_2 emissions as economies transition to a low-carbon energy future. Here, we have identified examples of key scientific and technological breakthroughs and challenges that are driving the capture and storage of CO_2, focusing on retrofits of coal combustion plants to highlight significant early implementation. We have also highlighted next-generation approaches that model how concerns of science, policy, and industry can be simultaneously addressed to make CCS a reality.

The last 10 years have seen significant improvements in all areas of CCS science, technology, and modeling, while industry investment is building a base of operational knowledge. The development of new technologies could reduce the cost of CO_2 capture to <\$20/tCO_2 within a decade (e.g., membrane technology). Current large geologic sequestration projects are now targeting injection rates of greater than 1 $MtCO_2$/yr, linked to industrial CO_2 sources. Even in the absence of federal carbon policy, companies like Southern Company, Denbury, SaskPower, and Ameren are building integrated CCS infrastructure for managing natural and anthropogenic CO_2 in large part from coal-fired power plants. The early CCS projects that focus on EOR benefits and public-private demonstration plants illustrate the benefits of integrated systems and economies of scale. At the same time, these projects highlight the precarious nature of costly

technology investments in the absence of national carbon economic policies. Independent of the progress on the scientific and engineering research side of this work, strong political will and public education will be required if we are to realize CCS on a scale that can neutralize anthropogenic impacts to the Earth's climate.

Major hurdles to success remain in all areas of CCS. In capture, the most promising new technologies are being demonstrated at the bench scale, and reaching target DOE costs for capture will require scaling these technologies up first to the pilot then the industrial scale. Scale is also a major issue for storage; for CCS to have a meaningful impact, the volumes of injected CO_2 will be considerably larger than anything previously attempted. For example, we will need to confirm that our current models of rock failure and fluid flow in fractures are appropriate when very large areas of the subsurface become pressurized with CO_2. We must address issues of liability and pore space ownership, where a growing patchwork of individual state regulations could limit the interest of industry to invest in CCS projects. Finally, scaling up CCS from pilot projects to a set of integrated networks nation-wide will require vision and planning. Lessons learned during early industrial-scale demonstrations can guide the development of CCS systems—capture technologies, dedicated pipelines, and storage reservoirs—that are capable of reliably and cost-effectively reducing CO_2 emissions on a meaningful scale.

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